

Amendments to the Claims

This listing will replace all prior versions and listings of claims in the application:

Listing of Claims

1. (original) A method of depositing a film of a metal chalcogenide, comprising the steps of:

contacting an isolated hydrazinium-based precursor of a metal chalcogenide and a solvent having therein a solubilizing additive to form a solution of a complex thereof;

applying said solution of said complex onto a substrate to produce a coating of said solution on said substrate;

removing said solvent from said coating to produce a film of said complex on said substrate; and thereafter

annealing said film of said complex to produce a metal chalcogenide film on said substrate.

2. (original) The method of claim 1, wherein said solvent is selected from the group consisting of: water, lower alcohol, ether, esters, alkylene glycol of 2-6 carbon atoms, dialkylene glycol of 4-6 carbon atoms, trialkylene glycol of 6 carbon atoms, glyme, diglyme, triglyme, propylene glycol monoacetate, DMSO, DMF, DMA, HMPA and a mixture thereof.

3. (original) The method of claim 2, wherein said solubilizing additive is selected from the group consisting of: an aliphatic amine of 1-10 carbon atoms, aromatic amine of 4-10 carbon atoms, aminoalcohol of 2-6 carbon atoms and a mixture thereof.

4. (original) The method of claim 2, wherein said solubilizing additive is selected from the group consisting of: n-propylamine, iso-propylamine, n-butylamine, sec-butylamine, iso-butylamine, pentylamine, n-hexylamine, cyclohexylamine, phenethylamine, pyridine, aniline, aminotoluene, ethanolamine, propanolamine, diethanolamine, dipropanolamine, triethanolamine, tripropanolamine and a mixture thereof.

5. (original) The method of claim 1, wherein said isolated hydrazinium-based precursor of said metal chalcogenide is prepared by a process comprising the steps of:

contacting: at least one metal chalcogenide, a hydrazine compound represented by the formula:



wherein each of R^1 , R^2 , R^3 and R^4 is independently selected from the group consisting of: hydrogen, aryl, methyl, ethyl and a linear, branched or cyclic alkyl of 3-6 carbon atoms, and optionally an elemental chalcogen selected from the group consisting of: S, Se, Te and a combination thereof, to produce a solution of a hydrazinium-based precursor of said metal chalcogenide in said hydrazine compound; and

isolating hydrazinium-based precursor of said metal chalcogenide as a substantially pure product.

6. (original) The method of claim 5, wherein said metal chalcogenide comprises a metal selected from the group consisting of: Ge, Sn, Pb, Sb, Bi, Ga, In, Tl and a combination thereof and a chalcogen selected from the group consisting of: S, Se, Te and a combination thereof.

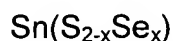
7. (original) The method of claim 5, wherein said metal chalcogenide is represented by the formula MX or MX_2 wherein M is a metal selected from the group consisting of: Ge, Sn, Pb and a combination thereof; and wherein X is a chalcogen selected from the group consisting of: S, Se, Te and a combination thereof.

8. (original) The method of claim 5, wherein said metal chalcogenide is represented by the formula M_2X_3 wherein M is a metal selected from the group consisting of: Sb, Bi, Ga, In and a combination thereof; and wherein X is a chalcogen selected from the group consisting of: S, Se, Te and a combination thereof.

9. (original) The method of claim 5, wherein said metal chalcogenide is represented by the formula M_2X wherein M is Tl; and wherein X is a chalcogen selected from the group consisting of: S, Se, Te and a combination thereof.

10. (original) The method of claim 5, wherein said metal is selected from the group consisting of: Sn and Sb; and wherein said chalcogen is selected from the group consisting of: S and Se.

11. (original) The method of claim 10, wherein said chalcogenide is represented by the formula:



wherein x is from 0 to 2.

12. (original) The method of claim 5, wherein each of R¹, R², R³ and R⁴ is independently selected from the group consisting of: hydrogen, aryl, methyl and ethyl.

13. (original) The method of claim 5, wherein R¹, R², R³ and R⁴ are hydrogen.

14. (original) The method of claim 5, wherein said metal chalcogenide film is in the form of a thin film.

15. (original) The method of claim 5, wherein said metal chalcogenide film comprises a polycrystalline metal chalcogenide or single crystals of said metal chalcogenide.

16. (original) The method of claim 15, wherein said polycrystalline metal chalcogenide has a grain size equal to or greater than the dimensions between contacts in a semiconductor device.

17. (original) The method of claim 5, wherein said annealing step is carried out at a temperature and for a length of time sufficient to produce said metal chalcogenide film.

18. (original) The method of claim 1, wherein said substrate is selected from the group consisting of:

Kapton, polycarbonate, silicon, amorphous hydrogenated silicon, silicon carbide (SiC), silicon dioxide (SiO₂), quartz, sapphire, glass, metal, diamond-like carbon, hydrogenated diamond-like carbon, gallium nitride, gallium arsenide, germanium, silicon-germanium, indium tin oxide, boron carbide, boron nitride, silicon nitride (Si₃N₄), alumina (Al₂O₃), cerium(IV) oxide (CeO₂), tin oxide (SnO₂), zinc titanate (ZnTiO₂), a plastic material and a combination thereof.

19. (original) A film prepared by the method of claim 5.

20. (original) The method of claim 1, wherein said hydrazinium-based precursor of said metal chalcogenide is prepared by a process comprising the steps of:

contacting: at least one metal chalcogenide and a salt of an amine compound with H₂S, H₂Se or H₂Te, wherein said amine compound is represented by the formula:



wherein each of R^5 , R^6 and R^7 is independently selected from the group consisting of: hydrogen, aryl, methyl, ethyl and a linear, branched or cyclic alkyl of 3-6 carbon atoms, to produce an ammonium-based precursor of said metal chalcogenide;

contacting said ammonium-based precursor of said metal chalcogenide, a hydrazine compound represented by the formula:



wherein each of R^1 , R^2 , R^3 and R^4 is independently selected from the group consisting of: hydrogen, aryl, methyl, ethyl and a linear, branched or cyclic alkyl of 3-6 carbon atoms, and optionally, an elemental chalcogen selected from the group consisting of: S, Se, Te and a combination thereof; to produce a solution of a hydrazinium-based precursor of said metal chalcogenide in said hydrazine compound; and

isolating hydrazinium-based precursor of said metal chalcogenide as a substantially pure product.

21. (original) The method of claim 20, wherein said metal chalcogenide comprises a metal selected from the group consisting of: Ge, Sn, Pb, Sb, Bi, Ga, In, Tl and a combination thereof and a chalcogen selected from the group consisting of: S, Se, Te and a combination thereof.

22. (original) The method of claim 20, wherein each of R^1 , R^2 , R^3 , R^4 , R^5 , R^6 and R^7 is independently selected from the group consisting of: hydrogen, aryl, methyl and ethyl.

23. (original) The method of claim 20, wherein R^1 , R^2 , R^3 , R^4 , R^5 , R^6 and R^7 are hydrogens.

24. (original) The method of claim 20, wherein said metal chalcogenide film is in the form of a thin film.

25. (original) The method of claim 20, wherein said metal chalcogenide film comprises a polycrystalline metal chalcogenide or single crystals of said metal chalcogenide.

26. (original) The method of claim 25, wherein said polycrystalline metal chalcogenide has a grain size equal to or greater than the dimensions between contacts in a semiconductor device.

27. (original) The method of claim 20, wherein said annealing step is carried out at a temperature and for a length of time sufficient to produce said metal chalcogenide film.

28. (original) The method of claim 20, wherein said substrate is selected from the group consisting of:

Kapton, polycarbonate, silicon, amorphous hydrogenated silicon, silicon carbide (SiC), silicon dioxide (SiO_2), quartz, sapphire, glass, metal,

diamond-like carbon, hydrogenated diamond-like carbon, gallium nitride, gallium arsenide, germanium, silicon-germanium, indium tin oxide, boron carbide, boron nitride, silicon nitride (Si_3N_4), alumina (Al_2O_3), cerium(IV) oxide (CeO_2), tin oxide (SnO_2), zinc titanate (ZnTiO_2), a plastic material and a combination thereof.

29. (original) A film prepared by the method of claim 20.

30. (original) A method of forming a field-effect transistor of the type having a source region and a drain region, a channel layer extending between the source region and the drain region, the channel layer including a semiconducting material, a gate region disposed in spaced adjacency to the channel layer, an electrically insulating layer between the gate region and the source region, drain region and channel layer, comprising:

preparing a channel layer comprising a film of a metal chalcogenide semiconducting material by a method comprising the steps of: contacting an isolated hydrazinium-based precursor of a metal chalcogenide and a solvent having therein a solubilizing additive to form a solution of a complex thereof; applying said solution of said complex onto a substrate to produce a coating of said solution on said substrate; removing said solvent from said coating to produce a film of said complex on said substrate; and thereafter annealing said film of said complex to produce a metal chalcogenide film on said substrate.

31. (original) The method of claim 30, wherein said source region, channel layer and drain region are disposed upon a surface of a substrate, said electrically insulating layer is disposed over said channel layer and

extending from said source region to said drain region, and said gate region is disposed over said electrically insulating layer.

32. (original) The method of claim 30, wherein said gate region is disposed as a gate layer upon a surface of a substrate, said electrically insulating layer is disposed upon said gate layer, and said source region, channel layer, and drain region are disposed upon said electrically insulating layer.

33. (original) The method of claim 30, wherein said metal chalcogenide film is in the form of a thin film having a thickness of from about 5 Å to about 2,000 Å.

34. (original) The method of claim 30, wherein said metal chalcogenide film comprises a polycrystalline metal chalcogenide or single crystals of said metal chalcogenide.

35. (original) The method of claim 34, wherein said metal chalcogenide film is polycrystalline with a grain size equal to or greater than the dimensions between contacts in a semiconductor device.

36. (original) The method of claim 30, wherein said metal chalcogenide comprises a metal selected from the group consisting of: Ge, Sn, Pb, Sb, Bi, Ga, In, Tl and a combination thereof and a chalcogen selected from the group consisting of: S, Se, Te and a combination thereof.

37. (original) The method of claim 30, wherein said annealing step is carried out at a temperature and for a length of time sufficient to produce said metal chalcogenide film.

38. (original) The method of claim 37, wherein said temperature is from about 25 °C to about 500 °C.

39. (original) The method of claim 38, wherein said temperature is from about 250 °C to about 350 °C.

40. (original) The method of claim 30, wherein said substrate is selected from the group consisting of:

Kapton, polycarbonate, silicon, amorphous hydrogenated silicon, silicon carbide (SiC), silicon dioxide (SiO₂), quartz, sapphire, glass, metal, diamond-like carbon, hydrogenated diamond-like carbon, gallium nitride, gallium arsenide, germanium, silicon-germanium, indium tin oxide, boron carbide, boron nitride, silicon nitride (Si₃N₄), alumina (Al₂O₃), cerium(IV) oxide (CeO₂), tin oxide (SnO₂), zinc titanate (ZnTiO₂), a plastic material and a combination thereof.

41. (original) A field-effect transistor prepared by the method of claim 30.

42. (original) A process for preparing an isolated hydrazinium-based precursor of a metal chalcogenide comprising the steps of:

contacting: at least one metal chalcogenide, a hydrazine compound represented by the formula:



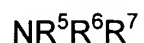
wherein each of R^1 , R^2 , R^3 and R^4 is independently selected from the group consisting of: hydrogen, aryl, methyl, ethyl and a linear, branched or cyclic alkyl of 3-6 carbon atoms, and optionally an elemental chalcogen selected from the group consisting of: S, Se, Te and a combination thereof, to produce a solution of a hydrazinium-based precursor of said metal chalcogenide in said hydrazine compound; and

isolating the hydrazinium-based precursor of said metal chalcogenide as a substantially pure product.

43. (original) A hydrazinium metal chalcogenide prepared by the process of claim 42.

44. (original) A process for preparing an isolated hydrazinium-based precursor of a metal chalcogenide comprising the steps of:

contacting: at least one metal chalcogenide and a salt of an amine compound with H_2S , H_2Se or H_2Te , wherein said amine compound is represented by the formula:



wherein each of R^5 , R^6 and R^7 is independently selected from the group consisting of: hydrogen, aryl, methyl, ethyl and a linear, branched or cyclic

alkyl of 3-6 carbon atoms, to produce an ammonium-based precursor of said metal chalcogenide;

contacting said ammonium-based precursor of said metal chalcogenide, a hydrazine compound represented by the formula:

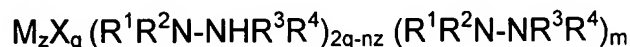


wherein each of R^1 , R^2 , R^3 and R^4 is independently selected from the group consisting of: hydrogen, aryl, methyl, ethyl and a linear, branched or cyclic alkyl of 3-6 carbon atoms, and optionally, an elemental chalcogen selected from the group consisting of: S, Se, Te and a combination thereof; to produce a solution of a hydrazinium-based precursor of said metal chalcogenide in said hydrazine compound; and

isolating hydrazinium-based precursor of said metal chalcogenide as a substantially pure product.

45. (original) A hydrazinium metal chalcogenide prepared by the process of claim 44.

46. (original) A hydrazinium metal chalcogenide represented by the formula:



wherein:

M is a main-group metal having a valence n, wherein n is an integer from 1 to 6;

X is a chalcogen;
z is an integer from 1 to 10;
q is an integer from 1 to 30;
m is from 1 to 30.5; and
each of R^1 , R^2 , R^3 and R^4 is independently selected from the group consisting of: hydrogen, aryl, methyl, ethyl and a linear, branched or cyclic alkyl of 3-6 carbon atoms.

47. (original) The hydrazinium metal chalcogenide of claim 46, wherein said metal is selected from the group consisting of: Ge, Sn, Pb, Sb, Bi, Ga, In and Tl.

48. (original) The hydrazinium metal chalcogenide of claim 46, wherein said chalcogen is selected from the group consisting of: S, Se and Te.

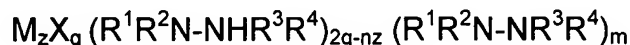
49. (original) A film prepared by the method of claim 1.

-- 50. (new) A method of preparing a multilayer film on a substrate, said multilayer film having a plurality of metal chalcogenide films; wherein each of said metal chalcogenide films is prepared by a method comprising the steps of:

- (a) contacting an isolated hydrazinium-based precursor of a metal chalcogenide and a solvent having therein a solubilizing additive to form a solution of a complex thereof;
- (b) applying said solution of said complex onto a substrate to produce a coating of said solution on said substrate;

- (c) removing said solvent from said coating to produce a film of said complex on said substrate; and thereafter
- (d) annealing said film of said complex to produce a metal chalcogenide film on said substrate; and
- (e) repeating steps (a), (b), (c) and (d) with the same or different chalcogenide to produce said multilayer film.

51. (new) The method of claim 50, wherein said isolated hydrazinium-based precursor is represented by the formula:



wherein:

M is a main-group metal having a valence n, wherein n is an integer from 1 to 6;

X is a chalcogen;

z is an integer from 1 to 10;

q is an integer from 1 to 30;

m is from 1 to 30.5; and

each of R^1 , R^2 , R^3 and R^4 is independently selected from the group consisting of: hydrogen, aryl, methyl, ethyl and a linear, branched or cyclic alkyl of 3-6 carbon atoms.

52. (new) The method of claim 50, wherein said metal is selected from the group consisting of: Ge, Sn, Pb, Sb, Bi, Ga, In and Tl.

53. (new) The method of claim 50, wherein said chalcogenide is selected from the group consisting of: S, Se and Te.

54. (new) The method of claim 50, wherein said multilayer film comprises a plurality of two alternating metal chalcogenide films each selected from the group consisting of: SnSe_2 , GeSe_2 , In_2Se_3 , Sb_2S_3 , and Sb_2Se_3 .

55. (new) The method of claim 50, wherein said metal chalcogenide film is produced at a temperature from about 25 °C to about 500 °C.

56. (new) The method of claim 50, wherein said substrate is selected from the group consisting of:

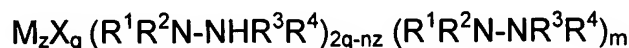
Kapton, silicon, amorphous hydrogenated silicon, silicon carbide (SiC), silicon dioxide (SiO_2), quartz, sapphire, glass, metal, diamond-like carbon, hydrogenated diamond-like carbon, gallium nitride, gallium arsenide, germanium, silicon-germanium, indium tin oxide, boron carbide, boron nitride, silicon nitride (Si_3N_4), alumina (Al_2O_3), cerium(IV) oxide (CeO_2), tin oxide (SnO_2), zinc titanate (ZnTiO_2), a plastic material and a combination thereof.

57. (new) A device comprising:

a multilayer active layer having a plurality of metal chalcogenide films; wherein each of said metal chalcogenite films is prepared by a method comprising steps of:

- (a) contacting an isolated hydrazinium-based precursor of a metal chalcogenide and a solvent having therein a solubilizing additive to form a solution of a complex thereof;
- (b) applying said solution of said complex onto a substrate to produce a coating of said solution on said substrate;
- (c) removing said solvent from said coating to produce a film of said complex on said substrate; and thereafter
- (d) annealing said film of said complex to produce a metal chalcogenide film on said substrate; and
- (e) repeating steps (a), (b), (c) and (d) with the same or different chalcogenide to produce said multilayer film.

58. (new) The device of claim 57, wherein said isolated hydrazinium-based precursor is represented by the formula:



wherein:

M is a main-group metal having a valence n, wherein n is an integer from 1 to 6;

X is a chalcogen;

z is an integer from 1 to 10;

q is an integer from 1 to 30;

m is from 1 to 30.5; and

each of R^1 , R^2 , R^3 and R^4 is independently selected from the group consisting of: hydrogen, aryl, methyl, ethyl and a linear, branched or cyclic alkyl of 3-6 carbon atoms.

59. (new) The device of claim 57, wherein said metal is selected from the group consisting of: Ge, Sn, Pb, Sb, Bi, Ga, In and Tl.

60. (new) The device of claim 57, wherein said chalcogenide is selected from the group consisting of: S, Se and Te.

61. (new) The device of claim 57, wherein said multilayer film comprises a plurality of two alternating metal chalcogenide films each selected from the group consisting of: SnSe₂, GeSe₂, In₂Se₃, Sb₂S₃, and Sb₂Se₃.

62. (new) The device of claim 57, wherein said metal chalcogenide film is produced at a temperature from about 25 °C to about 500 °C.

63. (new) The device of claim 57, wherein said substrate is selected from the group consisting of:

Kapton, silicon, amorphous hydrogenated silicon, silicon carbide (SiC), silicon dioxide (SiO₂), quartz, sapphire, glass, metal, diamond-like carbon, hydrogenated diamond-like carbon, gallium nitride, gallium arsenide, germanium, silicon-germanium, indium tin oxide, boron carbide, boron nitride, silicon nitride (Si₃N₄), alumina (Al₂O₃), cerium(IV) oxide (CeO₂), tin oxide (SnO₂), zinc titanate (ZnTiO₂), a plastic material and a combination thereof.

64. (new) The device of claim 57, wherein said device is selected from the group consisting of: flat panel displays, non-linear optical devices, non-

linear photo-conductive devices, chemical sensors, light-emitting diodes, thin-film transistors, field-effect transistors, media for optical data storage, phase change media for optical data storage, solar cells and thermoelectric devices.

65. (new) A method of preparing a multilayer film on a substrate, said multilayer film having a plurality of metal chalcogenide films; wherein each of said metal chalcogenide films is prepared by a method comprising steps of:

(a) contacting a metal chalcogenide and (i) a hydrazine compound, optionally in the presence of additional amount of an elemental chalcogen, or (ii) first a salt of an amine compound with H_2S , H_2Se or H_2Te , and thereafter a hydrazine compound, to produce a solution of a hydrazinium-based precursor of said metal chalcogenide;

(b) applying said solution of said hydrazinium-based precursor of said metal chalcogenide onto a substrate to produce a film of said precursor;

(c) annealing said film of said precursor under conditions sufficient to produce said metal chalcogenide film; and

(d) repeating steps (a), (b) and (c) with the same or different chalcogenide to produce said multilayer film.

66. (new) The method of claim 65, wherein said metal is selected from the group consisting of: Ge, Sn, Pb, Sb, Bi, Ga, In and Tl.

67. (new) The method of claim 65, wherein said chalcogenide is selected from the group consisting of: S, Se and Te.

68. (new) The method of claim 65, wherein said multilayer film comprises a plurality of two alternating metal chalcogenide films each

selected from the group consisting of: SnSe₂, GeSe₂, In₂Se₃, Sb₂S₃, and Sb₂Se₃.

69. (new) The method of claim 65, wherein said hydrazine compound is represented by the formula:



wherein each of R¹, R², R³ and R⁴ is independently selected from the group consisting of: hydrogen, aryl, methyl, ethyl and a linear, branched or cyclic alkyl of 3-6 carbon atoms; and wherein said amine compound is represented by the formula:



wherein each of R⁵, R⁶ and R⁷ is independently selected from the group consisting of: hydrogen, aryl, methyl, ethyl and a linear, branched or cyclic alkyl of 3-6 carbon atoms.

70. (new) The method of claim 65, wherein said metal chalcogenide film is produced at a temperature from about 25 °C to about 500 °C.

71. (new) The method of claim 65, wherein said substrate is selected from the group consisting of:

Kapton, silicon, amorphous hydrogenated silicon, silicon carbide (SiC), silicon dioxide (SiO₂), quartz, sapphire, glass, metal, diamond-like carbon,

hydrogenated diamond-like carbon, gallium nitride, gallium arsenide, germanium, silicon-germanium, indium tin oxide, boron carbide, boron nitride, silicon nitride (Si_3N_4), alumina (Al_2O_3), cerium(IV) oxide (CeO_2), tin oxide (SnO_2), zinc titanate (ZnTiO_2), a plastic material and a combination thereof.

72. (new) A device comprising:

a multilayer active layer having a plurality of metal chalcogenide films; wherein each of said metal chalcogenite films is prepared by a method comprising steps of:

(a) contacting a metal chalcogenide and (i) a hydrazine compound, optionally in the presence of additional amount of an elemental chalcogen, or (ii) first a salt of an amine compound with H_2S , H_2Se or H_2Te , and thereafter a hydrazine compound, to produce a solution of a hydrazinium-based precursor of said metal chalcogenide;

(b) applying said solution of said hydrazinium-based precursor of said metal chalcogenide onto a substrate to produce a film of said precursor;

(c) annealing said film of said precursor under conditions sufficient to produce said metal chalcogenide film; and

(d) repeating steps (a), (b) and (c) with the same or different chalcogenide to produce said multilayer active layer in said device.

73. (new) The device of claim 72, wherein said metal is selected from the group consisting of: Ge, Sn, Pb, Sb, Bi, Ga, In and Tl.

74. (new) The device of claim 72, wherein said chalcogenide is selected from the group consisting of: S, Se and Te.

75. (new) The device of claim 72, wherein said multilayer film comprises a plurality of two alternating metal chalcogenide films each selected from the group consisting of: SnSe_2 , GeSe_2 , In_2Se_3 , Sb_2S_3 , and Sb_2Se_3 .

76. (new) The device of claim 72, wherein said hydrazine compound is represented by the formula:



wherein each of R^1 , R^2 , R^3 and R^4 is independently selected from the group consisting of: hydrogen, aryl, methyl, ethyl and a linear, branched or cyclic alkyl of 3-6 carbon atoms; and wherein said amine compound is represented by the formula:



wherein each of R^5 , R^6 and R^7 is independently selected from the group consisting of: hydrogen, aryl, methyl, ethyl and a linear, branched or cyclic alkyl of 3-6 carbon atoms.

77. (new) The device of claim 72, wherein said metal chalcogenide film is produced at a temperature from about 25 °C to about 500 °C.

78. (new) The device of claim 72, wherein said substrate is selected from the group consisting of:

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Kapton, silicon, amorphous hydrogenated silicon, silicon carbide (SiC), silicon dioxide (SiO₂), quartz, sapphire, glass, metal, diamond-like carbon, hydrogenated diamond-like carbon, gallium nitride, gallium arsenide, germanium, silicon-germanium, indium tin oxide, boron carbide, boron nitride, silicon nitride (Si₃N₄), alumina (Al₂O₃), cerium(IV) oxide (CeO₂), tin oxide (SnO₂), zinc titanate (ZnTiO₂), a plastic material and a combination thereof.

79. (new) The device of claim 72, wherein said device is selected from the group consisting of: flat panel displays, non-linear optical devices, non-linear photo-conductive devices, chemical sensors, light-emitting diodes, thin-film transistors, field-effect transistors, media for optical data storage, phase change media for optical data storage, solar cells and thermoelectric devices.--